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# REPORT

on Item 0002 of the
Contract No. F61775-99-WE030 / 23.04.1999

## Study of productivity of tantalum spallation by protons for accumulation of the $^{178m2}$Hf isomeric nuclei

## I. Introduction

The goal of a gamma-ray laser is closer than ever to its realization. After many years of trials the first successful induced gamma emission from a high excitation energy isomeric state was reported in 1999 [1] and reconfirmed afterwards [2]. Many methods of producing an induced gamma ray emission were proposed (see Ref. [3] for review) but the first one leading to a successful result was the triggering of the energy stored in high-K nuclear isomeric state. It is the case of the 31 years K=16 isomer of $^{178}$Hf placed at 2.45 MeV excitation energy. The experiments were performed at the University of Texas at Dallas, the Center for Quantum Electronics by the group leaded by Prof. C. B. Collins and with the contribution of the IGE Foundation. We have to notice that this is just a first step towards the realization of the gamma ray laser and it provides an isotropicaly distributed gamma ray emission. Nevertheless, even such an emission could be of large applicability (see Ref. [4] for a review of the possible applications) thought that the gain in energy defined as the gamma-ray output energy divided by the X-ray input energy is more than two orders of magnitude. Another very important result of our experiment was the low energy of the X-rays needed to trigger the release of the gamma-ray energy stored in the long-lived isomeric state. As a consequence one can use small sized simple devices to produce the triggering radiation eliminating safety and portability related problems. The most relevant example is the experiment itself [1,2] that was performed with a simple X-ray unit normally used in dental examination.

Several problems related to the practical realization of the gamma-ray laser has still to be faced and the IGE Foundation, in parallel with the activity connected to the isomer triggering research, has started to investigate upon various options and solutions. One

can identify several phases for the building of a gamma-ray laser:

a) production of the nuclei in isomeric state in macroscopic quantities;

b) the induced emission of the energy stored in the isomeric states;

c) achieving coherency and monochromaticity of the emitted gamma-rays.

We have solved the second phase from the list above. For any further development one needs to figure out the way to produce large quantities of the material in isomeric sate. The available resources are extremely limited and they diminish continuously with the lifetime of the isomer and the number of successful triggering experiments. Consequently, the IGE Foundation has initiated a research direction on the large yield reactions producing $^{178}$Hf nuclei in isomeric state. The largest quantity of such isomeric nuclei has been produced in 1978 at LAMPF meson factory [5] at Los Alamos through spallation of the Ta beam dump by protons of 800 MeV energy. Production of $^{178}$Hf isomeric nuclei appeared as a by-product and consequently it was never optimized. Other methods for producing the isomeric $^{178}$Hf nuclei were identified (see Ref. [6] for details) but even if optimized, as it is the case of the $^{176}$Yb + $\alpha$ reaction used at the Dubna cyclotron, their production yield was lower than the spallation process. The production process of the isomeric nuclei has to be performed in several steps. First we have to elucidate which is the best mechanism to produce large amounts of isomeric nuclei. In other words: is the spallation the best mechanism for the isomer production?; if yes, which is the best target to be used? Then we have to deal with finding the best chemical and mass separation methods of the $^{178}$Hf isotope. One needs afterwards to handle the radioactive waste (storage, degradation) remaining after the separation of the isotope. In a more refined step one should face the problem of separating the isomeric nuclei from the ground state ones. Such separation is commonly used for low spin isomers with laser techniques but they are still not effective for high spin isomeric states.

The major aim of the present contract with EOARD was to answer the question if the spallation process can be really optimized to get a better yield of the $^{178}$Hf isomeric nuclei and if it is really superior to the other known mechanisms to produce isomeric $^{178}$Hf nuclei as concern the isomer yield, purity and cooling time of the final sample. It was also meant to calibrate the available spallation modeling programs to get

realistic estimates on the production yield so that the best target for spallation can be chosen. The first part of the contract was devoted to the experimental part, as reported in the Interim Report [6], while the second part was dedicated to calibrate the parameters of the computer codes with the experimental results from the spallation on Ta target and to make predictions for passing to a natural Re target.

## I I . Spallation - general remarks and modeling

Spallation reactions have a wide range of applications in different fields of activity as astrophysics, geophysics, radiotherapy, radiobiology, and nucleosynthesis. Sometime the process is called with a different term but, despite the non uniformity of the terminology, it is commonly accepted that spallation reactions are inelastic nuclear reactions in which at least one of the two collision partners is a complex nucleus and in which the energy available well exceeds the interaction energy between nucleons in the nucleus. Thus, a nucleon-nucleus or nucleus-nucleus collision in which the incident energy exceeds 50 or 100 MeV/amu is generally referred to as a spallation reaction. The term come from the verb 'to spall' meaning to chip with a hammer which is a very suggestive image of the way the incident nucleon or nucleus 'spall' away from the nucleus several nucleons.

Within a simple model, spallation reactions are described as a two-step process. In the first step the high-energy particle enters the target and interacts with some of the individual nucleons from the target nucleus. Consequently, a few nucleons will be knocked-out from the target nucleus. The residual target nucleus with less nucleons is left in an excited state and it de-excites in the second step of the model. The nucleus de-excites through particle evaporation (protons, neutrons, alpha etc.) and gamma-rays leading to the final nucleus in isomeric or ground state. The first step is called as the 'fast' stage, the 'knock-out' stage or, more commonly, the 'cascade' stage. The second step is called as the 'slow' stage, the 'de-excitation' stage or simply the 'evaporation' stage. One have to consider the fact that usually the irradiated targets are thick so that the nucleons expelled by the primary beam from the target nucleus induce further interactions in the target giving rise to secondary spallation process and so on until their energy decrease below the cut-off energy. This is referred as 'nuclear

nuclear cascade' in the target or 'selfmultiplying spallation' reactions.

A big improvement in the calculation of the spallation processes was achieved when Monte Carlo and transport techniques were used for the simulations. The computer program we used for the calculations is one of the best that can be found on the market nowadays and is based on the techniques mentioned above. The code is called LAHET [7] and was developed based on two previously used codes: LANL and HETC [8]. The code LAHET treats the spallation reactions in a more refined way considering them as three step processes. The first step is the nuclear cascade as discussed above. After this first step, a preequilibrium emission phase can be considered. This stage corresponds to a transitional regime from the direct reaction to the statistical decay of the compound nucleus. At each stage of this regime the excited nucleus may emit nuclear particles as the nucleonic configuration evolves towards equilibrium. The last step corresponds to the evaporation. The computer code describes this step by taking in account the competition between the evaporation of nuclear particles and fission.

Generally, the computer codes calculate the cumulative yield for one type of nuclei regardless they have or not isomeric states. It is for the first time that we tried to optimize the calculations from this point of view.

## I I I . Experimental results

The first part of the present contract was devoted to the experimental testing of the possibility to optimize the Ta spallation by high-energy protons. The results were reported in the Interim Report [5] and here we remind only the main conclusions of the report.

Prior to our experimental work the only available data for the spallation of Ta were provided following the irradiation with 800 MeV proton beam at LAMPF [5]. We considerably extended the knowledge on this process by performing irradiation with protons at three other different energies: 100, 200 and 660 MeV, respectively, covering a large range of energies. The analysis was focused on finding the best

irradiation energy that ensures a high enough absolute production rate of the isomeric $^{178m2}$Hf nuclei and a low enough degree of contamination of the final sample.

Our analysis leaded to several very important conclusions for the continuation of isomer production by spallation with high-energy protons.

a) Spallation at lower proton energies is more convenient due to the lower cost of the production process, the availability of several accelerators around the world able to deliver the proton beams and a lower contamination with $^{172}$Hf, as well;

b) The use of thin targets prevents the accumulation and production of undesired contaminants; this results in a lower level of activity of the sample and, consequently, in a shortening of its cooling time;

c) Thin targets prevent heating problems and no special cooling systems should be placed around them;

d) The targets should be changed periodically to prevent the accumulation of $^{178}$Hf nuclei in the ground state and to keep a higher isomer-to-ground state ratio;

Fig. 1 shows the evolution in time of the concentration of $^{178m2}$Hf nuclei and the main contaminant, $^{172}$Hf, in the samples produced by spallation of the Ta targets at different proton beam energies.
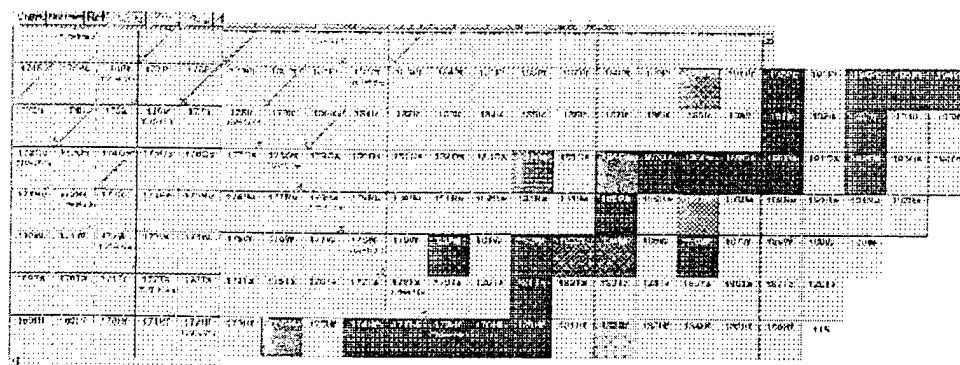


*Figure 1. Partial nuclide chart showing the isotopes neighboring $^{178}$Hf. The goal of our studies is to maximize the production yield of $^{178m2}$Hf isomeric nuclei and to minimize the access to the path decaying to the principal long-lived contaminant, $^{172}$Hf. Arrows indicate known decay chains leading to the elements of concern.*

In the case of Ta spallation by high-energy protons we showed that at 100 MeV beam energy we get a reduction of the cooling time of the samples from 20 years in the case

of the samples resulted at LAMPF to about 6-7 years. Also, the contamination with $^{172}$Hf is reduced by order of magnitudes.

We estimated that by using a 150 $\mu$A proton beam at 100 MeV energy in one year of continuous irradiation one can get about 50 $\mu$g of $^{178m2}$Hf nuclei which is about 50 times more than the production through $\alpha$ particles induced reactions.

## I V . New perspectives for $^{178m2}$Hf production through spallation by high-energy protons

The analysis that we shortly outlined above has clearly shown that the production of milligrams of $^{178m2}$Hf isomeric nuclei is still a goal to be achieved.

Generally, the spallation process favors the population of nuclei through many particles (neutrons, protons, $\alpha$ particles) evaporation. In the case of $^{181}$Ta spallation by protons the $^{178m2}$Hf nuclei appear following the evaporation of 2 protons and 2 neutrons. The relative yield for such a process involving only a few particles evaporation is as low as 0.03% of the total yield. A way to increase the production yield of $^{178m2}$Hf nuclei is to change to a heavier target type as Re or W which would require a larger number of particles to be evaporated for the population of the final nucleus. The question was: which would be the best candidate for such a target. The target material should satisfy a few requirements as: a) it has to be available in large quantities in nature to keep as low as possible the target production costs; b) the spallation of the target has to be performed at moderate high energies to ensure the easy access to accelerators that can provide such proton beams; c) the isotopic composition of the natural target should be reduced as much as possible to avoid spallation on many different isotopes. Fig. 2 shows a map of the isotopes heavier than Ta and the various paths that lead to $^{178}$Hf nuclei by spallation with protons. One of the most promising material to be used as target is Re. The new experimental data available from our measurements [6] provided us with valuable information needed to calibrate the parameters of the computer codes used to simulate the spallation processes.

# Spallation Results
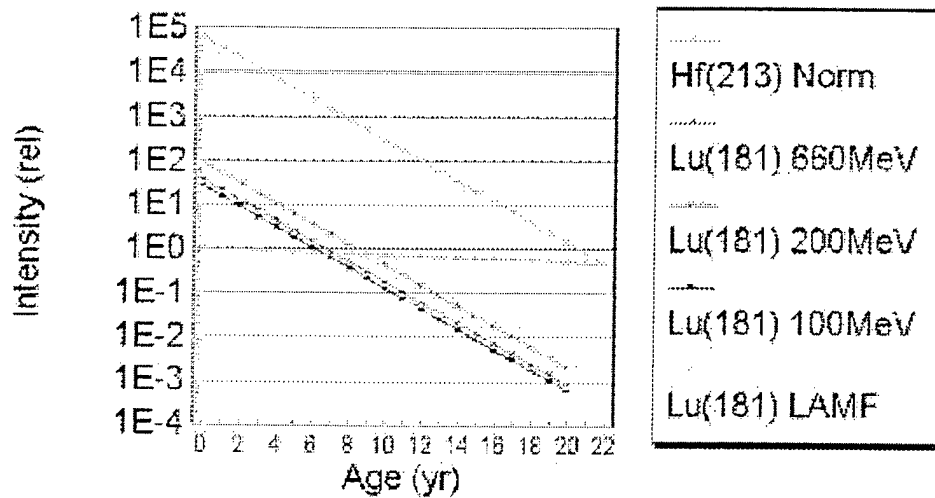
## IGE Foundation for EOARD



*Figure 2. A useful figure-of-merit that reflects the ratio of isomer to contaminant is aging of the product material necessary for the latter to decay down to the level at which the relative intensities of the (2,4) transitions of the respective ground state bands are the same. It can be seen that the curves of intensity as a function of age cross in only 6 years for the product produced in our irradiations in comparison to 22 years for the LAMPF product.*

We calculated the p + $^{181}$Ta system with the LAHET code for the three cases corresponding to the three irradiation energies, 100, 200 and 660 MeV, respectively. A very complete analysis could be performed for the 660 MeV. The irradiation at 660 MeV populated a large number of nuclei covering the whole range of possible cases resulted after spallation. The analysis of the experimental data has revealed as many as 35 isotopes and isomers populated at this energy. Their yields, from independent or cumulated population after the beta-decay or electron capture of the parent nuclei, were measured down to cross-sections of the order of 10 $\mu$barn. This case represented a real ground test for the ability of the computer code to reproduce the experimental results and revealed its limits for quantitative predictions. The calculations were performed with 5 million events for each one of the energies. This allowed us to calculate nuclei produced with yields down to $3 \times 10^{-7}$. The comparison experiment-theory is presented in Table 1.

**Table 1.** *Yields of nuclei and mean cross-sections measured after activation of a 33.3 g/cm² thickness Ta target by 660 MeV protons [6]. The comparison with theory prediction is given at the last column.*

| Nuclide | T$_{1/2}$ | Type of yield | Yield value *) atoms/proton | Mean σ, mbarn | Y$_{exp}$/Y$_{theor}$ |
|---|---|---|---|---|---|
| $^{182}$Ta | 115 d | Indep. | $0.85 \cdot 10^{-3}$ | - | - |
| $^{181}$Hf | 42.4 d | Indep. | $1.7 \cdot 10^{-5}$ | 0.14 | 0.60 |
| $^{179m2}$Hf | 25.1 d | Indep. | $5.8 \cdot 10^{-5}$ | 0.52 | - |
| $^{178}$W | 21.6 d | Indep. | $6.6 \cdot 10^{-4}$ | 5.9 | 0.71 |
| $^{178m2}$Hf | 31 y | Indep. | $3.5 \cdot 10^{-5}$ | 0.31 | - |
| $^{177m}$Lu | 161 d | Indep. | $2.8 \cdot 10^{-5}$ | 0.25 | - |
| $^{175}$Hf | 70 d | EC cum. | $6.2 \cdot 10^{-3}$ | 56 | 1.26 |
| $^{174g}$Lu | 3.31 y | Indep. | $1.7 \cdot 10^{-4}$ | 1.5 | } 1.46 |
| $^{174m}$Lu | 142 d | Indep. | $5.3 \cdot 10^{-4}$ | 4.8 | |
| $^{173}$Lu | 1.37 y | EC cum. | $7.7 \cdot 10^{-3}$ | 70 | 1.42 |
| $^{172}$Hf | 1.87 y | EC cum. | $5.2 \cdot 10^{-3}$ | 47 | 1.13 |
| $^{172}$Lu | 6.7 d | Indep. | $0.9 \cdot 10^{-3}$ | 8.1 | 1.07 |
| $^{171}$Lu | 8.22 d | EC cum. | $7.8 \cdot 10^{-3}$ | 71 | 1.44 |
| $^{170}$Lu | 2.0 d | EC cum. | $7.1 \cdot 10^{-3}$ | 64 | 1.33 |
| $^{169}$Yb | 32.0 d | EC cum. | $8.3 \cdot 10^{-3}$ | 75 | 1.51 |
| $^{168}$Tm | 93.1 d | Indep. | $1.5 \cdot 10^{-4}$ | 1.3 | 2.12 |
| $^{167}$Tm | 9.24 d | EC cum. | $7.9 \cdot 10^{-3}$ | 72 | 1.62 |
| $^{166}$Dy | 3.4 d | β⁻ cum. | $2.0 \cdot 10^{-4}$ | 1.8 | no theory |
| $^{160}$Tb | 72.3 d | Indep. | $1.2 \cdot 10^{-5}$ | 0.10 | no theory |
| $^{156}$Tb | 5.35 d | Indep. | $5.3 \cdot 10^{-5}$ | 0.48 | 1.15 |
| $^{156}$Eu | 15.2 d | β⁻ cum. | $1.3 \cdot 10^{-5}$ | 0.12 | no theory |
| $^{149}$Gd | 9.4 d | EC cum. | $3.0 \cdot 10^{-3}$ | 27 | 1.73 |
| $^{149}$Eu | 93.1 d | EC cum. | $3.1 \cdot 10^{-3}$ | 28 | 1.67 |
| $^{148}$Eu | 54.3 d | Indep. | $0.8 \cdot 10^{-4}$ | 0.73 | 0.5 |
| $^{148g}$Pm | 5.37 d | Indep. | $5 \cdot 10^{-6}$ | 0.045 | } 12.5 |
| $^{148m}$Pm | 41.3 d | Indep. | $7.5 \cdot 10^{-6}$ | 0.067 | |
| $^{147}$Eu | 24.6 d | EC cum. | $1.68 \cdot 10^{-3}$ | 15 | 1.11 |
| $^{146}$Gd | 48.3 d | EC cum. | $1.54 \cdot 10^{-3}$ | 14 | 1.40 |
| $^{146}$Eu | 4.6 d | Indep. | $5.4 \cdot 10^{-5}$ | 0.5 | 0.15 |
| $^{145}$Eu | 5.94 d | EC cum. | $0.94 \cdot 10^{-3}$ | 8.5 | 0.88 |
| $^{144}$Pm | 363 d | Indep. | $\leq 2 \cdot 10^{-4}$ | $\leq 1.7$ | in accordance |
| $^{143}$Pm | 265 d | EC cum. | $6.5 \cdot 10^{-4}$ | 5.8 | 0.73 |
| $^{140}$Ba | 12.75 d | β⁻ cum. | $1.2 \cdot 10^{-6}$ | 0.010 | no theory |
| $^{136}$Cs | 13.16 d | Indep. | $\leq 1.3 \cdot 10^{-6}$ | $\leq 0.011$ | no theory |

*) Random errors are within ±7%, and the standard error of the absolute calibration is of about ±15%.

We have to mention that the computer code is not able to calculate separately the population of isomers and gives only the total yield for a given isotope disregarding the division among ground state and isomer(s). Fig. 3a shows the mass distribution

resulted after the spallation of $^{181}$Ta by 660 MeV protons compared to the calculations.
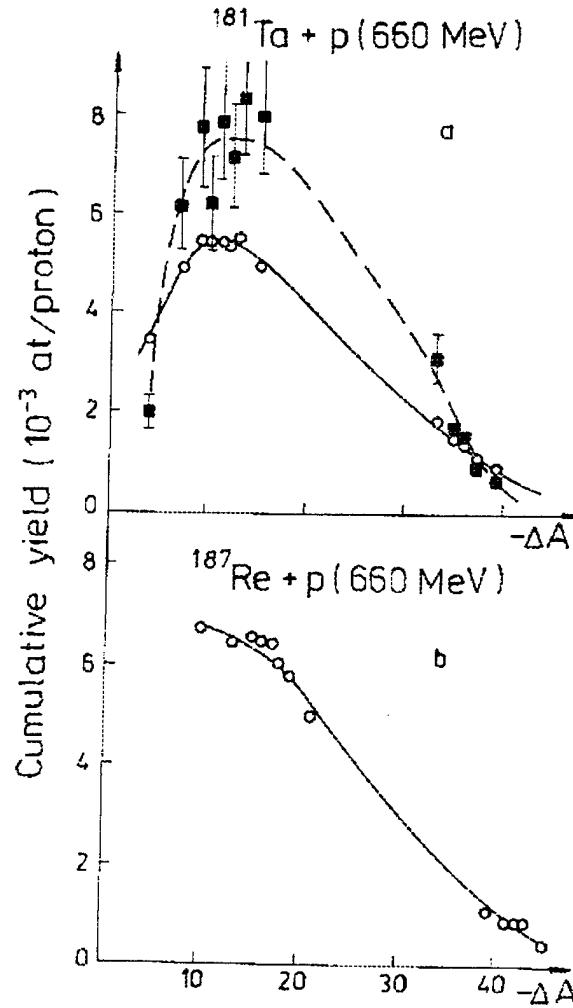


**Figure 3.** *Mass distributions of the cumulative yields for nuclei produced through the spallation of a)* $^{181}$Ta *and b)* $^{187}$Re *by 660 MeV protons. Filled squares represent the experimental data while the opened circles the calculated values. Lines are drawn only to guide the eye. By -ΔA we mean the quantity $A_t+1-A_p$ where $A_t$ is the atomic mass of the target and $A_p$ is the atomic mass of the product nucleus. It measures the number of particles emitted during the reaction.*

From the analysis of Table 1 and Fig. 3 one can immediately notice that for most of the nuclei we have a very good agreement experiment-theory to within a factor 2. We can use this factor to empirically correct the predictions for other systems, as p + $^{nat}$Re The yields for the neutron-rich isotopes are underestimated by the calculations even

by order of magnitudes (see the case of $^{166}$Dy, $^{160}$Tb, $^{148}$Pm, $^{140}$Ba). The direct yield for nuclei that have the Z/A ratio far from the most probable values are again only poorly reproduced and the actual values could be much higher than the calculated ones. This is the case of $^{178}$Hf production through $^{nat}$Re spallation with protons. In the case of isomers we are interested in the direct yield since this is the only one that can produce nuclei in isomeric states. Any other process that contributes to the cumulative yield will just produce more nuclei in the ground state and consequently will degrade the isomer-to-ground-state ratio.

*Table 2. Calculated by the LAHET code yield of radioisotopes in the irradiation of $^{nat}$Re target by 660 MeV protons in the geometry the same as in the experiment with Ta target*

| Nuclide | $T_{1/2}$ | Type of yield | Yield value, at./proton |
|---|---|---|---|
| $^{181}$Hf | 42.4 d | Indep. | $3.33 \cdot 10^{-5}$ |
| $^{179}$Hf | Stable + isomer | Indep. | $7.93 \cdot 10^{-5}$ |
| $^{178}$W | 21.6 d | EC cum. | $5.87 \cdot 10^{-3}$ |
| $^{178}$Hf | Stable | EC cum. | $6.80 \cdot 10^{-3}$ |
| $^{178}$Hf | Stable + isomer | Indep. | $1.16 \cdot 10^{-4}$ |
| $^{177}$Lu | 6.7 d + 161 d | Indep. | $1.27 \cdot 10^{-5}$ |
| $^{175}$Hf | 70 d | EC cum. | $6.47 \cdot 10^{-3}$ |
| $^{174}$Lu | 3.31 y + 142 d | Indep. | $8.20 \cdot 10^{-5}$ |
| $^{173}$Lu | 1.37 y | EC cum. | $6.57 \cdot 10^{-3}$ |
| $^{172}$Hf | 1.87 y | EC cum. | $6.21 \cdot 10^{-3}$ |
| $^{172}$Lu | 6.7 d | Indep. | $2.95 \cdot 10^{-4}$ |
| $^{171}$Lu | 8.22 d | EC cum. | $6.41 \cdot 10^{-3}$ |
| $^{170}$Lu | 2.0 d | EC cum. | $6.05 \cdot 10^{-3}$ |
| $^{169}$Yb | 32.0 d | EC cum. | $5.82 \cdot 10^{-3}$ |
| $^{168}$Tm | 93.1 d | Indep. | $3.0 \cdot 10^{-5}$ |
| $^{167}$Tm | 9.24 d | EC cum. | $5.15 \cdot 10^{-3}$ |
| $^{156}$Tb | 5.35 d | Indep. | $3.2 \cdot 10^{-5}$ |
| $^{149}$Gd | 9.4 d | EC cum. | $1.16 \cdot 10^{-3}$ |
| $^{149}$Eu | 93.1 d | Indep. | $8.80 \cdot 10^{-5}$ |
| $^{148}$Eu | 54.3 d | Indep. | $1.12 \cdot 10^{-4}$ |
| $^{148}$Pm | 5.37 d + 41.3 d | Indep. | $6.67 \cdot 10^{-7}$ |
| $^{147}$Eu | 24.6 d | EC cum. | $9.70 \cdot 10^{-4}$ |
| $^{146}$Gd | 48.3 d | EC cum. | $7.08 \cdot 10^{-4}$ |
| $^{146}$Eu | 4.6 d | Indep. | $2.13 \cdot 10^{-4}$ |
| $^{145}$Eu | 5.94 d | EC cum. | $6.39 \cdot 10^{-4}$ |
| $^{144}$Pm | 363 d | Indep. | $3.60 \cdot 10^{-5}$ |
| $^{143}$Pm | 265 d | EC cum. | $4.76 \cdot 10^{-4}$ |

The p + $^{nat}$Re system was calculated under the same conditions as the p + $^{181}$Ta one. The $^{nat}$Re target was considered to consist of the most abundant two isotopes $^{185}$Re and $^{187}$Re. The calculations at the lower proton energies, 100 and 200 MeV, respectively, have not yielded significant amounts of $^{178}$Hf and these cases were ruled out from the analysis. The only case of interest was at 660 MeV and the results of the calculations are summarized in Table 2.
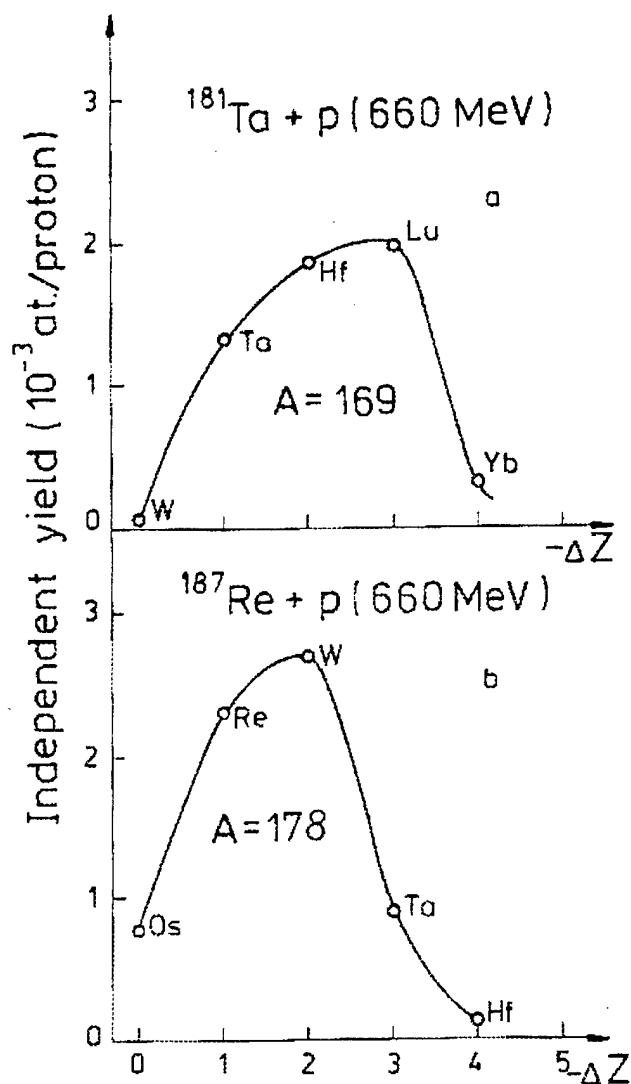


*Figure 4. Isobaric charge distribution for a) A=169 nuclei produced from the spallation of 181Ta by 660 MeV protons and b) A=178 nuclei resulting after the spallation of 187Re by 660 MeV protons as calculated by the LAHET computer code.*

Fig. 3b shows the mass distribution of the nuclei populated through the spallation of $^{187}$Re at 660 MeV. It can be seen that mass 178 (corresponding to a -$\Delta$A=10) is now shifted to the maximum of the distribution. The A=178 isobars relative direct yield distribution as a function of the Z number is shown in Fig. 4b. The $^{178}$Hf nucleus is clearly not on the maximum but as we stressed above its independent yield can be underestimated even by one order of magnitude. The $^{178}$W nucleus has a higher yield and through electron capture it populates the ground state of $^{178}$Hf. So, one should take care to eliminate the $^{178}$W contribution from the irradiated samples before it decays out in a significant amount.

Another unknown problem related to the isomer production is the isomer-to-ground-state ratio inside the estimated independent yield for the $^{178}$Hf nucleus. This is a problem that was only qualitatively discussed in previous papers due to the limits of the models employed to describe the process. These limits are related to the knowledge of the angular momentum distribution for the reaction residues and the isomer-to-ground-state ratio behavior for a given angular momentum distribution. Since the theory cannot yet face this problem the straightest manner to treat it is a semiempiric one based on the experimentally measured results. The study of the Ta spallation by protons at different incident energies provided us information on two isomers of $^{174}$Lu and $^{148}$Pm. In these two nuclei the isomeric states and the ground state are only independently populated and we can get an accurate estimate of the isomer-to-ground-state ratio. For the two nuclei the ratio is higher than unity and has an increasing tendency towards higher bombarding energies. One can consider a simple model to calculate these ratios that would consider the primary momentum transfer at the collision moment and the emission of particles. After the emission of particles the angular momentum $I_f$ is defined by the primary transferred angular momentum and the angular momentum removed by the emitted particles. Further de-excitation of the nucleus through gamma decay and internal electron conversion will bring the nucleus to the isomeric or ground state. The initial angular momentum $I_i$ depends on the impact parameters and the $\Delta$A value. In the case of p + $^{181}$Ta at 660 MeV we have a distribution of $I_i$ with a width of about (10 - 20) $\hbar$. For a given $\Delta$A value he angular momentum distribution is relatively narrow. In the case considered above for $\Delta$A=10 we have an initial distribution of the angular momentum of about

(15 - 20) ħ. Each emitted particle carry away an angular momentum of about $|\Delta l| = 3$ ħ. Since the $\Delta l$ vectors are randomly oriented we will get a broadening of the angular momentum distribution. For the p + $^{181}$Ta we get a distribution of the final angular momenta from 0 to 25 ħ. The distributions of the angular momenta discussed above are shown in Fig. 5.
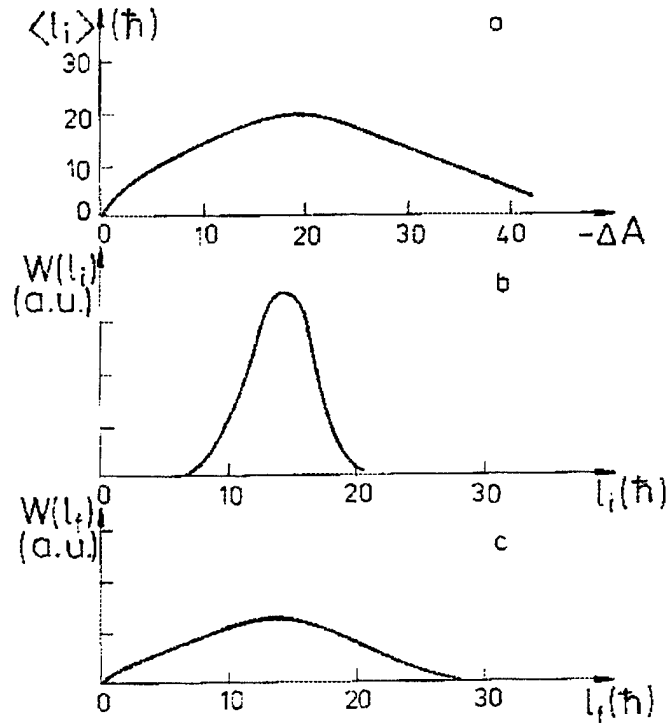


*Figure 5. Estimation of the angular momentum distribution during the spallation process. a) distribution of the mean angular momentum transferred in the first stage of the reaction; b) the angular momentum distribution for given value -ΔA=10, after the first step of the reaction; c) the same distribution as the one from the previous point but after the evaporation stage.*

The final angular momentum distribution for mass A=178 (corresponding to ΔA=10) in the case of the p + $^{187}$Re at 660 MeV bombarding energy should look like the one in Fig. 5c. Then one can evaluate the direct population isomer-to-ground-state ratio to be about (0.2 - 0.3) for the $16^+$ $^{178m2}$Hf isomeric state.

We have to notice that in the case of nuclei populated after few particle emissions the calculations show that the isomer-to-ground-state ratio is much lower than the unity.

This is the case of the $^{178m2}$Hf isomer populated through the Ta spallation ($\Delta A=4$). An increase of the ratio is obtained for nuclei populated through many particles evaporation; this is the case of $^{178}$Hf when populated through the spallation of Re by protons ($-\Delta A=10$).


**Conclusions**

The present contract was aimed to investigate the possibilities to optimize the spallation process in order to get a higher yield for the $^{178m2}$Hf isomeric nuclei. The first part was devoted to the study of the Ta spallation by high energy protons at three different bombarding energies, 100, 200 and 660 MeV, respectively. The results obtained have shown that the process can be optimized to increase the production of $^{178m2}$Hf nuclei but the production of milligrams of such material is still far away if Ta target are used. The experimental work has proven to be of very big importance for the modeling of the spallation processes by high energy protons and allowed us to calibrate the computer codes for making an investigation of other systems that can lead to $^{178m2}$Hf nuclei through spallation by protons. As spallation favors nuclei populated through many nucleons emission we considered other materials, heavier than Tantalum, as targets. The most promising materials look to be $^{187}$Re and $^{186}$W. Enriched targets of these materials are very expensive and for the next round of experimental measurements we thought to use natural Rhenium composed mainly of $^{185}$Re and $^{187}$Re. The use of Tungsten as target would have produced supplementary difficulties concerning the chemical separation of Hf and W. Calculations show that the optimum bombarding energy is 660 MeV. The calculated yield for $^{178}$Hf is of the same order as in the case of Ta spallation but this is just a lower limit in the case of Re and it could be even by an order of magnitude higher. The contamination of the sample with $^{172}$Hf and $^{175}$Hf remains at the same level as in the case of Ta spallation. The Re targets should be changed at periodically in order to prevent $^{178}$Hf in ground state accumulation following the decay of the $^{178}$W nuclei. Within a simple model one could estimate that the isomer-to-ground-state ratio for the case of Re spallation should be much higher than in the case of Ta spallation.

By combining all the results listed above one can conclude that Re spallation by high energy protons is much more better than the Ta spallation yielding a higher quantity of $^{178m2}$Hf isomers and a better isomer-to-ground-state ratio at the same level of population of the main contaminants. The results of the calculations will be extremely useful for guiding us during the next round of experimental measurements for the Re spallation by protons.

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